Atmospheric iron deposition and sea-surface dissolved iron concentrations in the eastern Atlantic Ocean

Sarthou, Géraldine; Baker, Alex R.; Blain, Stéphane; Achterberg, Eric P.; Boye, Marie; Bowie, Andrew R.; Croot, Peter; Laan, Patrick; Baar, Hein J.W. de; Jickells, Timothy D.

Published in:
Deep-Sea Research I

DOI:
10.1016/S0967-0637(03)00126-2

Publication date:
2003

Available online at www.sciencedirect.com
SCIENCE DIRECT®
PERGAMON
Deep-Sea Research I 50 (2003) 1339–1352
DEEP-SEA RESEARCH PART I
www.elsevier.com/locate/dsr

Atmospheric iron deposition and sea-surface dissolved iron concentrations in the eastern Atlantic Ocean

Géraldine Sarthou[a,*], Alex R. Baker[b], Stéphane Blain[a], Eric P. Achterberg[c], Marie Boye[d], Andrew R. Bowie[c], Peter Croot[d], Patrick Laan[d], Hein J.W. de Baar[d], Timothy D. Jickells[b], Paul J. Worsfold[c]

[a] *Universite de Bretagne Occidentale, LEMAR/UMR 6539/IUEM, Technopole Brest Iroise Place Nicolas Copernic, F-29280 Plouzané, France*
[b] *School of Environmental Sciences, University of East Anglia, Norwich NR4 7TJ, UK*
[c] *Department of Environmental Sciences, University of Plymouth, Drake Circus, Plymouth PL4 8AA, UK*
[d] *Royal Netherlands Institute for Sea Research (NIOZ), Postbus 59, NL-1790 AB Den Burg Texel, The Netherlands*

Received 11 October 2002; received in revised form 26 June 2003; accepted 8 July 2003

Abstract

Atmospheric iron and underway sea-surface dissolved (<0.2 μm) iron (DFe) concentrations were investigated along a north–south transect in the eastern Atlantic Ocean (27°N/16°W–19°S/5°E). Fe concentrations in aerosols and dry deposition fluxes of soluble Fe were at least two orders of magnitude higher in the Saharan dust plume than at the equator or at the extreme south of the transect. A weaker source of atmospheric Fe was also observed in the South Atlantic, possibly originating in southern Africa via the north-easterly outflow of the Angolan plume. Estimations of total atmospheric deposition fluxes (dry plus wet) of soluble Fe suggested that wet deposition dominated in the intertropical convergence zone, due to the very high amount of precipitation and to the fact that a substantial part of Fe was delivered in dissolved form. On the other hand, dry deposition dominated in the other regions of the transect (73–97%), where rainfall rates were much lower. Underway sea-surface DFe concentrations ranged 0.02–1.1 nM. Such low values (0.02 nM) are reported for the first time in the Atlantic Ocean and may be (co)-limiting for primary production. A significant correlation (Spearman's rho = 0.862, $p < 0.01$) was observed between mean DFe concentrations and total atmospheric deposition fluxes, confirming the importance of atmospheric deposition on the iron cycle in the Atlantic. Residence time of DFe in the surface waters relative to atmospheric inputs were estimated in the northern part of our study area (17 ± 8 to 28 ± 16 d). These values confirmed the rapid removal of Fe from the surface waters, possibly by colloidal aggregation.

Keywords: Dissolved iron; Aerosol iron; Sea surface; Iron supply; Atmospheric (wet and dry) deposition; Residence time

*Corresponding author. Tel: +33-2-98-49-86-73; fax: +33-2-98-49-86-45.
E-mail address: geraldine.sarthou@univ-brest.fr (G. Sarthou).

0967-0637/$ - see front matter © 2003 Elsevier Ltd. All rights reserved.
doi:10.1016/S0967-0637(03)00126-2

1. Introduction

Iron (Fe) is an essential micro-nutrient for phytoplankton growth and is now known to be a limiting factor for primary productivity in large areas of the world ocean (Martin and Fitzwater, 1988; Martin et al., 1990, 1994; Coale et al., 1996; Boyd et al., 2000; Smetacek, 2001). Understanding the systematic delivery of Fe to surface waters is of great interest since it is in short supply due to its high insolubility and reactivity (de Baar and de Jong, 2001). The atmosphere plays an important role in material transport from land to sea and may represent the dominant external source of trace elements, such as Fe, to most of the remote surface ocean (Duce et al., 1991; Duce and Tindale, 1991; Jickells and Spokes, 2001). Dust production, transport and deposition processes are highly temporally variable. For example, in the Mediterranean Sea, events of high magnitude and short duration account for a large part of the annual input (Loÿe-Pilot and Martin, 1996), and at Midway in the central Pacific and over the Atlantic Ocean, half of the annual deposition occurs in 2 and 10 weeks, respectively (Prospero et al., 1989; Swap et al., 1996). In the Canary region, dust inputs lasting an average of 3–8 days are produced during winter and summer periods (Torres-Padron et al., 2002). Aeolian inputs are also highly spatially variable: about half the global aeolian input enters the North Pacific Ocean, originating from mainland Asia, and another third is deposited into the North Atlantic from the Sahara (Duce and Tindale, 1991; Jickells and Spokes, 2001). Layers of dust from the Sahara desert are entrained into the atmosphere at heights of 1–5 km (Carlson and Prospero, 1972; Maring et al., 2000) and transported westwards under the prevailing weather systems (Moulin et al., 1997). Dust concentrations over the South Atlantic are much lower than over the (sub)-tropical North Atlantic (Völkening and Heumann, 1990; Losno et al., 1992; Rädlein and Heumann, 1992; Bates et al., 2001), owing to much weaker dust sources. Atmospheric dust inputs occur by either wet or dry deposition. They typically result in an enrichment of surface Fe concentrations (Bruland et al., 1994; Wu and Luther III, 1994; Sarthou and Jeandel, 2001), since crustal materials are Fe-rich (~4.3%, by weight, Wedepohl, 1995). Nevertheless, the solubility of Fe in atmospheric particles is extremely low (Jickells and Spokes, 2001) and the residence time of Fe in surface seawater is rather short (Jickells, 1999; de Baar and de Jong, 2001) so that in some regions with high atmospheric inputs such as the Mediterranean Sea, after spring blooms and in the absence of any Saharan dust input, Fe concentrations in the surface waters are extremely low and may be limiting for phytoplankton growth (<0.1 nM; Sarthou and Jeandel, 2001).

Here we present results of atmospheric and surface seawater sampling aboard R.V. *Polarstern* during cruise ANTXVIII/1, in October 2000, in the eastern Atlantic Ocean (27°N–20°S, Fig. 1). The aim of this study was to investigate dissolved iron (DFe) concentrations in the surface waters in relation to atmospheric Fe deposition fluxes in

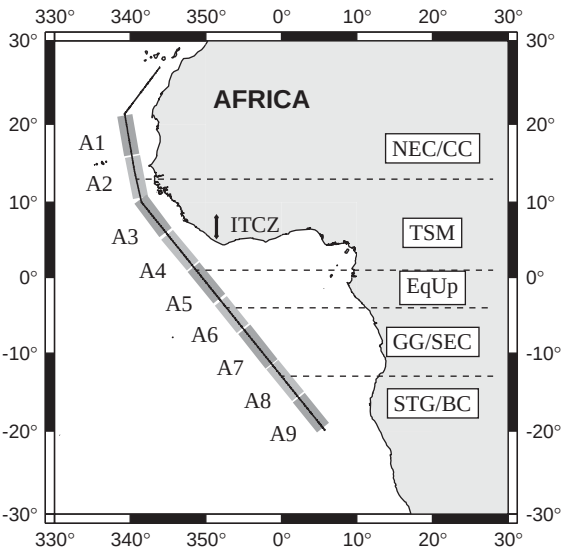

Fig. 1. Ship's track during cruise ANTXVIII/1 on-board R.V. *Polarstern* (29 September–23 October 2000). The shaded rectangles represent the aerosol samplings. The various oceanic regimes are reported: NEC/CC = North Equatorial Current and Canary Current; TSM = Tropical Salinity Minimum; EqUp = Equatorial Upwelling; GG/SEC = Guinea Gyre and South Equatorial Current; STG/BG = South Tropical Gyre and BC. The Intertropical Convergence Zone (ITCZ) is indicated with the black arrow.

order to better understand the input and removal processes controlling the Fe cycle.

2. Sampling and methods

2.1. Aerosol and rain samples

Aerosol samples for Fe analysis were collected with a high-volume ($1\,m^3\,min^{-1}$) sampler (Graseby–Anderson), between 21.1°N and 19.6°S. Aerosol collection was not possible between 27°N and 21.1°N, as the ship's course coincided with the prevailing wind direction, so that exhaust from the ship's stack was blown over the collector during this period. The sample collection period was ~23 h, and the sampler was fitted with a cascade impactor head in order to achieve size fractionation of the aerosol particles. Results here are reported for a coarse/fine size split of 1 μm, as this differentiates between mechanically generated (coarse) particles and particles created by gas-to-particle conversion processes (Raes et al., 2000). The upper stages of the impactor contained acid-washed, quartz fibre slotted filters and the back-up stage contained an acid-washed cellulose filter (Whatman 41). Details of sample collection are given in Table 1. During our cruise, a single rain event was sampled (7.9°N, 16.8°W) on passage through the intertropical convergence zone (ITCZ). The sample (~35 ml) was collected using an acid-washed polypropylene funnel (28 cm diameter) and acid-washed low-density polyethylene bottle.

Soluble Fe was extracted from aerosol samples with ammonium acetate buffer (pH 4.7) as a model for Fe release from aerosol in rainwater. This procedure is similar to the method employed by Bruland et al. (2001) to indicate the presence of bioavailable Fe in upwelling particulate matter off the coast of California. Portions of each filter were cut into small pieces with acid-washed, ceramic-bladed scissors and suspended in 25 ml of buffer solution for 1–2 h, after which the supernatant was filtered (0.2 μm, cellulose acetate filter) and acidified with Aristar concentrated HNO_3 to 0.4% v/v HNO_3. Fe was determined in the extracts by graphite furnace atomic absorption spectrometry (GFAAS). Filter blanks were determined with unexposed, acid-washed filters and cross-checked with unexposed portions of sample filters. Procedural blanks, in which filter papers were loaded into sampling cassettes for ~24 h but with no air drawn through them, showed no significant increase in concentration. Concentrations in extracts were converted into atmospheric concentrations by calculating the total quantity of Fe on each filter, after appropriate blank correction, and dividing by the known volume of air filtered for each sample. Uncertainties quoted for atmospheric concentrations (and deposition fluxes) have been calculated from standard deviations of replicate analyses and propagated for each stage of data analysis (blank subtraction, etc.) by standard error propagation methods. Aliquots of rain were filtered immediately after collection through 0.2 μm cellulose acetate filters, and both filtered

Table 1
Collection times, positions and air volumes for Fe aerosol samples

Sample	Date and time[a] (UTC)	Position[a]	Air volume (m^3)
A1	10/10/00, 14:05 11/10/00, 12:08	21.1°N 20.7°W 16.1°N 19.8°W	1317.9
A2	11/10/00, 13:12 12/10/00, 12:17	15.9°N 19.7°W 10.6°N 18.6°W	1487.2
A3	12/10/00, 13:00 12/10/00, 12:17	10.4°N 18.6°W 6.0°N 15.3°W	1487.4
A4	13/10/00, 13:45 14/10/00, 12:10	5.7°N 15.1°W 1.6°N 11.7°W	1359.9
A5	14/10/00, 13:04 15/10/00, 12:12	1.4°N 11.6°W 2.8°S 8.2°W	1441.7
A6	15/10/00, 14:05 16/10/00, 09:55	3.1°S 8.0°W 6.7°S 5.1°W	1173.0
A7	16/10/00, 11:30 17/10/00, 12:10	6.9°S 4.9°W 11.4°S 1.3°W	1470.9
A8	17/10/00, 13:00 18/10/00, 12:15	11.5°S 1.2°W 15.6°S 2.2°E	1416.3
A9	18/10/00, 13:10 19/10/00, 12:10	15.8°S 2.3°E 19.6°S 5.5°E	1364.2

[a] Upper values denote sampling start, lower values sampling end.

and unfiltered portions were acidified with Aristar concentrated HCl (10 µl per 10 ml). Rainwater Fe was determined by GFAAS. Funnel and filtration blanks were determined for the rain event and were found to be below the detection limit (5 nM).

2.2. Seawater samples

Underway water samples were collected between 27°N and 19°S. Sea-surface samples were collected with a fish towed from a winch extended ca. 5 m from the ship's starboard side and deployed at 1–2 m below the surface (de Jong et al., 1998; Bowie et al., 2002, in press). Surface water was pumped on-board with a peristaltic pump directly into a clean container and filtered on-line through a 0.2 µm filtration cartridge (Sartobran-P capsules, Sartorius). Samples were acidified to pH 2.0 with 1 ml of ultrapur® hydrochloric acid (HCl, Merck) per 1 l of sample for at least 1 h before analysis.

DFe concentrations were measured according to a chemiluminescence method adapted from Obata et al. (1993) (see Bucciarelli et al., 2001). The purification of the luminol solution through a column of 8-hydroxyquinoline immobilised on hydrophilic vinyl polymer (TSK-8-HQ after Diers-sen et al., 2001) improved both the blank and detection limit of the method. The blank was determined each day as the mean of five measure-ments of a low iron concentration sample with only 5 s of preconcentration and varied between 0.02 and 0.09 nM with an mean value of 0.06 ± 0.02 nM ($n = 19$). The detection limit, equal to three times the standard deviation of the blank, ranged 0.02–0.06 nM with an average value of 0.03 ± 0.01 nM. The individual contributions to the total blank of hydrochloric acid, ammonia, and ammonium acetate buffer were determined by addition of increasing amounts of these reagents to the sample and were lower than our detection limit.

3. Results

3.1. Air masses and hydrography

The air masses sampled were classified accord-ing to 5-day back trajectories calculated for arrival heights of 10, 500 and 1000 m above the ship's position (NOAA Air Resources Laboratory, HYSPLIT model, FNL data set). The northern end of the transect (>8°N) was characterised by north-easterly surface winds and high-level trajec-tories carrying dust off Saharan Africa (Fig. 2a). The coarse fractions of the first three aerosol samples in the transect all showed the orange/red colour characteristic of this dust. Marine air was then sampled during passage through the ITCZ (Fig. 2b). South of the equator, surface winds were from the south east (Fig. 2c and d), with 1000 m trajectories arriving from southern Africa around 5–10°S (Fig. 2c).

Distributions of sea-surface temperature (T) and salinity (S) were acquired on-board R.V. *Polarstern* (PODAS data acquisition system, ThermoSalinograph Bow, Fig. 3a and b). Chloro-phyll *a* (Chl *a*) data (Fig. 3c) were extracted from SEAWIFS images (GAC level 2, browse) obtained from the Ocean Colour Data website[1]. Images were reprocessed with SeaDAS (Baith et al., 2001) on a SUN Ultra-10 at NIOZ. Daily satellite passes through the study region were combined into single composite images for each day. These daily composites were then combined and averaged to produce the final image. T, S, and Chl *a* data allowed us to identify the typical oceanographic regimes encountered during the expedition.

North of 13°N, the ship's track passed through the North Equatorial Current and Canary Current (NEC/CC), characterised by lower temperatures and higher Chl *a* concentrations due to the North West African Upwelling. The wind systems of the northern and southern hemispheres converge in the ITCZ, producing heavy rain shower activity and an associated Tropical Salinity Minimum (TSM). The position of the ITCZ varies season-ally; it is close to the equator during northern winter and moves northward in northern summer with an abrupt shift occurring in late June (Siedler et al., 1992; Sultan and Janicot, 2000). During the cruise, rain shower activity typical of the ITCZ was located between 5.5°N and 8.3°N and the TSM was observed between 1°N and 13°N. South

[1] http://daac.gsfc.nasa.gov/data/dataset/SEAWIFS/index.html.

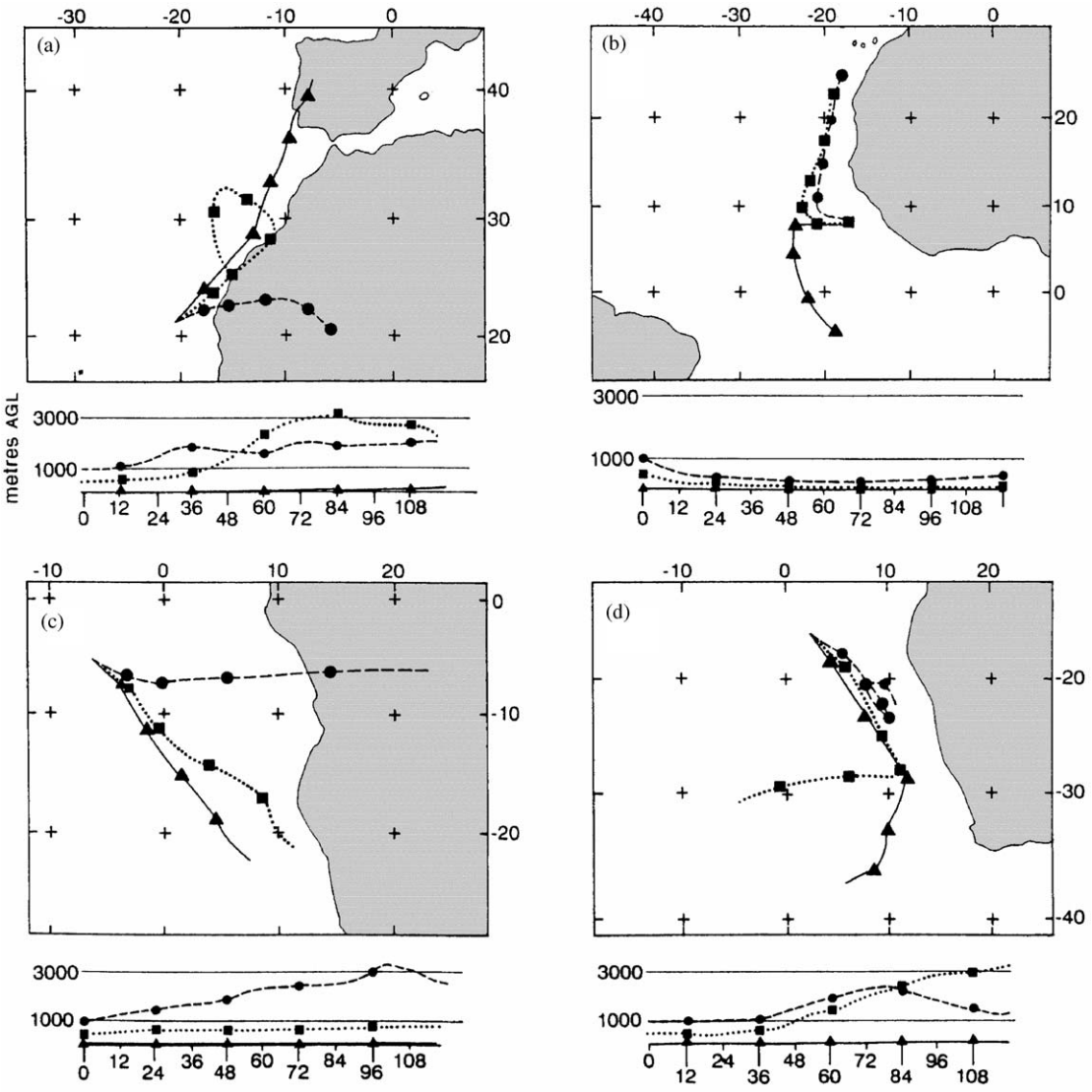

Fig. 2. Five-day air mass back trajectories for selected points along the ANTXVIII/1 cruise track: (a) 21.1°N 20.7°W, 10/10/00, 14:00, (b) 8.3°N 17.1°W, 13/10/00, 00:00, (c) 5.1°S 6.4°W, 16/10/00, 01:00, and (d) 15.7°S 2.3°E, 18/10/00, 13:00. Back trajectories converge at heights of 10 (triangles), 500 (squares) and 1000 m (circles) above the ship's position at the times indicated above. Lower sections of each panel show variation in height (m above ground level—AGL) of each air parcel with time (h) before arrival at ship's position.

of the TSM, the Equatorial Upwelling (EqUp, 1°N–4°S) was indicated by a lowered surface-water temperature and a small increase in surface-water salinity. Between 4°S and 13°S, the vessel crossed the Guinea Gyre/South Equatorial Current (GG/SEC) characterised by southward advection of low-salinity equatorial waters. Around 13°S, a temperature decrease was observed across the Angola/Benguela front (A/BF). This also marked the surface expression of the boundary between the GG to the north and the sub-tropical gyre (STG) to the south, which has the Benguela Current (BC) at its eastern boundary.

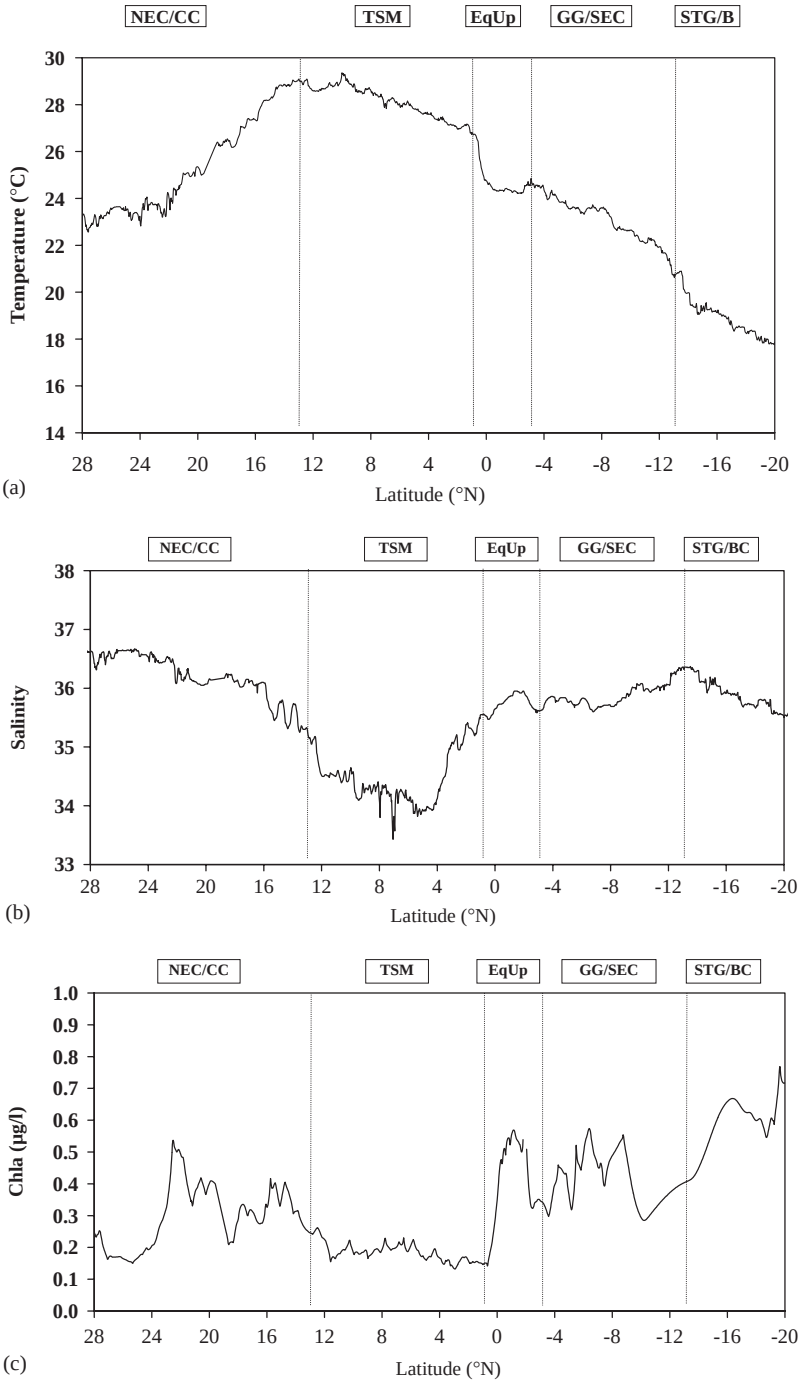

Fig. 3. (a) Temperature (°C) versus latitude (°N) (from PODAS underway system, R.V. *Polarstern*), (b) Salinity versus latitude (°N) (from PODAS underway system, R.V. *Polarstern*), and (c) Chl *a* (µg/l) versus latitude (°N) (extracted from SeaWIFS data). The oceanic regimes are separated by dotted lines.

3.2. Aerosol and rain iron

Soluble aerosol Fe concentrations ranged from 8 ± 2 to 199 ± 7 pmol m^{-3} for the fine particles and from 1 ± 1 to 512 ± 8 pmol m^{-3} for the coarse particles (Fig. 4). Concentrations were one–two orders of magnitude higher in the Saharan dust plume than at the equator or the southern extreme of the transect. A much smaller peak in soluble Fe concentrations occurred at $\sim 10°$S and coincided with a high-level air mass back trajectory from southern Africa (Fig. 2c). Aerosols associated with these high-level trajectories are likely to have originated in the north-easterly outflow of the Angolan plume (Tyson et al., 1996), where Fe is introduced principally from mineral dust (Piketh et al., 1999), but also possibly from biomass burning (Maenhaut et al., 1996). Soluble aerosol Fe concentrations were significantly lower than the total aerosol Fe values reported in previous studies (4–6.5 pmol m^{-3}: Völkening and Heumann, 1990; Losno et al., 1992; Rädlein and Heumann, 1992, 1995; Johansen et al., 2000), but probably correspond to total Fe concentrations of similar magnitude given that the solubility of aerosol Fe at the pH used for these extractions (pH 4.7) is likely to be of the order of 0.3–2% (Hoffmann et al., 1996; Spokes and Jickells, 1996; Johansen et al., 2000). The concentrations of Fe in the rainwater sample collected in the ITCZ were

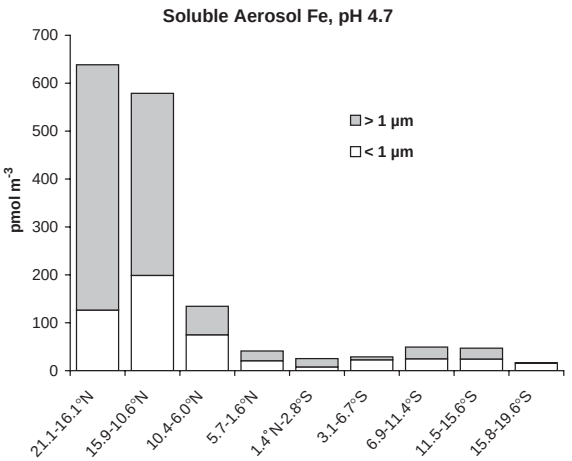

Fig. 4. Soluble (pH 4.7) iron concentrations in fine and coarse (shaded) mode aerosol.

280 ± 29 and 134 ± 6 nM for the unfiltered and the filtered (< 0.2 µm) fractions, respectively. These are within the range of values previously reported for unfiltered ITCZ rainwater samples in the Atlantic Ocean (150–1137 nM, Helmers and Schrem, 1995; volume weighted mean 530 nM, Kim and Church, 2002).

3.3. Sea-surface dissolved iron concentrations

During the cruise, a shipboard analytical intercomparison of DFe was performed by four different groups using two different flow injections-chemiluminescence methods (FI-CL) and competitive ligand equilibration-cathodic stripping voltammetry (CLE-CSV). Results of the intercomparison study are reported elsewhere (Bowie et al., in press). Briefly, the overall replicate precision ($2 \times$ SD) among investigators ranged 0.15–0.48 nM and was generally better than the 95% confidence intervals reported for the NASS Certified Reference Materials, which were 0.29 and 0.63 nM for NASS-4 and NASS-5, respectively. However, statistical analyses (paired two-tailed *t*-test) showed that each analytical method gave significantly different DFe concentrations at the 95% confidence interval. Nevertheless, when similar techniques were employed (FI-CL/University of Bretagne Occidentale (UBO) and FI-CL/Royal Netherlands Institute for Sea Research (NIOZ), see Bowie et al., in press), mean values were not significantly different at the 99% confidence interval ($P = 0.01$). In this paper, only one data set (FI-CL/UBO) is presented for internal consistency.

Along the north–south transect, DFe concentrations ranged from values as low as 0.02–1.1 nM (Fig. 5.). The average value along the whole transect was 0.28 ± 0.21 nM ($n = 194$). DFe concentrations were notably higher in the North than in the South Atlantic, with average values of 0.41 ± 0.19 ($n = 107$) and 0.11 ± 0.07 nM ($n = 85$), respectively. The maximum DFe concentration (1.11 nM) was observed in the northern part of NEC/CC around 28°N. South of 28°N, concentrations decreased sharply to reach 0.2–0.3 nM at 20°N. In the southern part of the NEC/CC, DFe increased again to 0.4–0.5 nM. In the TSM, DFe

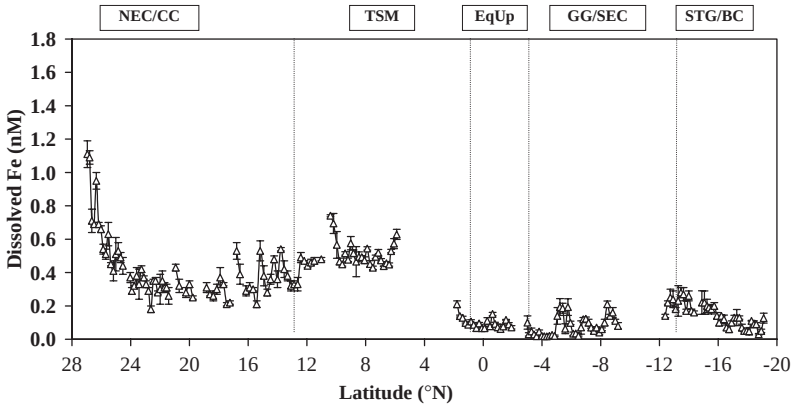

Fig. 5. Dissolved Fe (nM) versus latitude (°N).

averaged 0.50 ± 0.08 nM ($n = 33$) with values as high as 0.74 nM at 10.4°N. In the EqUp zone, DFe was very low, averaging values of less than 0.1 nM, and reached values as low as 0.02 nM south of this upwelling region. Finally, in the GG/SEC and STG/BC zones, DFe remained very low, ranging 0.02–0.28 nM and averaging 0.12 ± 0.08 nM ($n = 39$).

DFe data reported here are in general agreement with values reported along transects through the North and South Atlantic (Table 2), although this is the first time that such low values (0.02 nM) have been observed in the Atlantic Ocean. Vink and Measures (2001) observed sea-surface DFe concentrations along a transect in the south-west Atlantic ranging 0.4–1.4 nM, with a typical value of about 0.6–0.8 nM. In the equatorial Atlantic Ocean, Powell and Donat (2001) measured sea-surface DFe values varying between 0.28 and 0.71 nM with an averaged value of 0.55 ± 0.14 nM. Along a transect in the east Atlantic similar to our north–south transect, Powell et al. (1995) observed values ranging 0.5–10 nM for unfiltered samples. Finally, Bowie et al. (2002) reported concentrations between 0.3 and 4.5 nM and between 0.3 and 3.0 nM (samples also unfiltered) along two separate north–south transects of the Atlantic. Since the latter two studies were performed on unfiltered samples (i.e. total dissolvable Fe, TDFe), higher iron concentrations are expected due to partial dissolution of suspended particulate material.

4. Discussion

Dry deposition fluxes of soluble Fe aerosol were estimated as the sum of coarse (>1 μm) and fine (<1 μm) mode deposition:

$$F_{\mathrm{dry}} = C_{\mathrm{AFe}}\, v_{\mathrm{d}}, \tag{1}$$

where C_{AFe} is the concentration of Fe in each size mode, and v_{d} is the appropriate deposition velocity ($0.001\ \mathrm{m\,s^{-1}}$, fine mode; $0.02\ \mathrm{m\,s^{-1}}$, coarse mode; Duce et al., 1991).

The deposition velocity estimation is a first approximation since velocities are poorly constrained and uncertainties may be as much as a factor 2–3 (Duce et al., 1991). Moreover, soluble Fe was extracted from aerosol samples with ammonium acetate buffer (pH 4.7) and may not represent the dissolution processes that occur in seawater. Indeed, the solubility of iron is related strongly to pH and, at pH 4.7, Fe is more soluble than at seawater pH (Spokes et al., 1994). On the other hand, the higher ionic strength of seawater (Liu and Millero, 2002) and the presence of organic ligands increase Fe solubility (Gledhill and Van den Berg, 1994; Rue and Bruland, 1995, 1997). Interestingly, Powell and Donat (2001) have reported a 0.5 nM excess of natural Fe binding ligands in the surface waters of the equatorial North Atlantic Ocean, which may enhance Fe aerosol solubility.

Dry deposition fluxes varied between 3 ± 2 (15.8–19.6°S) and $896 \pm 14\ \mathrm{nmol\,m^{-2}\,d^{-1}}$ (21.1–

Table 2
Fe concentrations in Atlantic surface waters along north-south transects

Latitude/longitude	Depth range (m)	[DFe] range (nM)	[TDFe] range (nM)	[DFe] mean value (nM)	[TDFe] mean value (nM)	Number of data points	References
33°N–30°S/23°W–10°E	0.5–1		0.5–1.0			42	Powell et al. (1995)
50°N–52°S/4–57°W	<200 m		0.3–3.0		0.95±0.67	142	Bowie et al. (2002)
50N°–34°S/4°W–18°E	<200 m		0.3–4.5		1.08±0.59	160	Bowie et al. (2002)
5°N–16°S/23–45°W	~1	0.4–1.4					Vink and Measures (2001)
5°N–1°S/23–45°W	~1	0.28–0.71		0.55±0.14		10	Powell and Donat (2001)
27°N–19°S/21°W–4°E	~1	0.02–1.11		0.28±0.21		193	This study

DFe—dissolved Fe; TDFe—total dissolvable Fe.

16.1°N) (Fig. 6). As with aerosol Fe and sea-surface DFe concentrations, the lowest values were observed in the South Atlantic and were one–two orders of magnitude lower than in the Saharan dust plume.

Wet deposition is estimated globally to represent 30% of the total iron input to the ocean (Duce et al., 1991; Jickells and Spokes, 2001). However, in the ITCZ, rainfall can be as high as 2 m yr^{-1} (Tchernia, 1980; Elliott and Reed, 1984), and wet Fe deposition estimated in this zone (Helmers and Schrems, 1995) was similar to dry deposition in the western Atlantic (Lim and Jickells, 1990). In the ITCZ, wet deposition was shown to strongly influence the trace metal budget and the chemistry of surface seawater (Helmers and Schrems, 1995). In this region, we observed enhanced sea-surface DFe concentrations (Fig. 5), with maximum values of 0.74 nM. Vink and Measures (2001) also reported maximum values of DFe up to 1.4 nM, and TDFe concentrations as high as 3.7 and 10 nM were observed (Powell et al., 1995; Bowie et al., 2002, respectively). The Fe concentration in the filtered rain sample we collected in the ITCZ at 8°N (134±6 nM) was more than two orders of

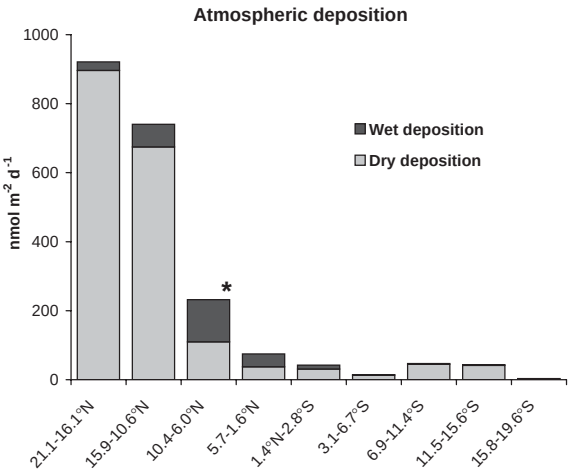

Fig. 6. Dry deposition fluxes (grey bars) of soluble Fe and wet deposition fluxes (black bars) of soluble Fe estimated from Fe concentration in aerosol samples and a scavenging ratio of 200, versus latitude. Dry deposition is dominated (>97%) by coarse mode aerosol. *Wet deposition of total Fe in the ITCZ was estimated from rainwater Fe concentrations to be 2200 nmol m^{-2} d^{-1}. See text for further details.

magnitude higher than the sea-surface DFe concentrations. Elevated total Fe concentrations (150–1100 nM) were also observed in rainwater samples in this region by Helmers and Schrems (1995), representing values 300–2300 times higher than DFe concentrations.

We estimated the wet deposition flux of Fe in the ITCZ according to

$$F_{wet} = C_{PFe}\, P, \quad (2)$$

where C_{PFe} is the concentration of Fe in the rainwater and P the precipitation rate ($2\,m\,yr^{-1}$). Given the large range of Fe concentrations in ITCZ rainwater samples, we use the volume-weighted mean total Fe concentration for our sample and those reported in Helmers and Schrem (1995) as a more representative value for C_{PFe} than our single sample alone. (Calculation of the volume-weighted mean is the mathematical equivalent of combining the samples and determining the concentration of the mixture.) The wet deposition flux estimated was $2200\,nmol\,m^{-2}\,d^{-1}$, which is very similar to the value of $2900\,nmol\,m^{-2}\,d^{-1}$ estimated by Kim and Church (2002) using a similar approach. Although the wet deposition flux of soluble Fe will be lower than this, it is still likely to be an order of magnitude higher than our estimated dry soluble Fe deposition flux in this area ($109 \pm 8\,nmol\,m^{-2}\,d^{-1}$), so that wet deposition dominates the atmospheric Fe flux in the ITCZ.

Although annual rainfall in the ITCZ is high, this rainfall typically takes the form of intense, isolated showers, so that collection of rain samples during passage through the ITCZ cannot be guaranteed (Helmers and Schrems, 1995). In other regions of our transect, annual rainfall is much lower (Elliott and Reed, 1984) and rain collection is therefore much more challenging. No samples were collected outside of the ITCZ during our cruise, nor have we been able to locate any literature data along our transect. In such circumstances, it is necessary to estimate concentrations in precipitation (C_P) from those in the aerosol phase (C_A) through the use of scavenging ratios (S_r, see Jickells and Spokes, 2001).

$$C_P = C_A S r/\rho, \quad (3)$$

where ρ is the density of air ($1200\,g\,m^{-3}$). The use of scavenging ratios has been criticised (Barrie, 1992) because rain forms at a different height from the measurements of aerosol concentrations. In the most recent estimate of dust inputs (Duce et al., 1991), scavenging ratios of 200 and 1000 were used for the Atlantic and the Pacific, respectively. Assuming a constant scavenging ratio of 200 along the transect and precipitation rates estimated by Elliot and Reed (1984), wet deposition fluxes of soluble Fe varied between 0.7 ± 0.1 (15.8–19.6°S) and $122 \pm 5\,nmol\,m^{-2}\,d^{-1}$ (10.4–6.0°N) (see Fig. 6). There is a clear discrepancy between our wet (total) Fe flux derived from rainfall data in the ITCZ (above) and the scavenging ratio-derived value for aerosol sample A3 (which included our passage through the ITCZ). This arises as a result of the very intense gradient in aerosol Fe concentration encountered for this sample (Saharan dust in the north, clean southern hemisphere air in the south), and because collection of the aerosol sample extended over a latitude range greater than that covered by the ITCZ, so that the two estimates do not refer to the same geographical region. This illustrates the difficulty of establishing atmospheric deposition fluxes in such a dynamic environment and we have therefore excluded sample A3 from the following discussion on the relationship between atmospheric Fe deposition and water column DFe concentrations.

Our estimates of total (wet + dry) deposition suggest that wet deposition dominates in the ITCZ, as observed by Helmers and Schrems (1995), whereas dry deposition dominates in all the other parts of the transect (73–97% of total deposition). A study of dust loading in the Canary Islands showed that dry deposition accounted for more than 80% of total (Torres-Padron et al., 2002), which is in agreement with our results in the northern part of the transect when influenced by the Saharan plume and in the southern part when influenced by the Angola plume. Wet deposition dominates in the ITCZ due to the very high amount of precipitation in this region. Moreover, the low pH values of rain, in combination with photochemical and organic complexation processes occurring in rainwater samples, ensure that a substantial part of Fe is delivered to the surface

ocean in dissolved form (Spokes et al., 1994; Spokes and Jickells, 1996; Zhu et al., 1993; Grgic et al., 1998). In our study, Fe concentration in the filtered rain fraction represented 48% of the Fe concentration in the unfiltered one.

DFe concentrations had a trend similar to atmospheric deposition fluxes of soluble Fe estimated from the aerosol phase, with higher values in the north (Fig. 7). While the distribution of our data was unsuitable to test for a linear relationship between these parameters, rank correlation analysis indicated that DFe concentrations and total soluble Fe deposition fluxes were positively correlated in this region of the Atlantic Ocean (Spearman's rho = 0.862, $p < 0.01$). Johnson et al. (1997), using measured and modelled values of DFe concentrations and Fe atmospheric fluxes, observed that DFe concentrations in the surface layer were uncorrelated with atmospheric fluxes. Nevertheless, in regions with high atmospheric fluxes, DFe may accumulate in the surface waters in some periods of the year: high sea-surface concentrations were observed during summer in the western Atlantic (0.58 nM, Wu and Luther III, 1994), in the central North Pacific (0.37 nM, Bruland et al., 1994), and in the Mediterranean Sea (4.8 nM, Sarthou and Jeandel, 2001; 2.72 nM, Guieu et al., 2002). On the other hand, iron deposited in the surface waters may be rapidly removed. During the bloom period in the Mediterranean Sea, the surface mixed layer can become Fe-depleted with values lower than 0.1 nM (Sarthou and Jeandel, 2001).

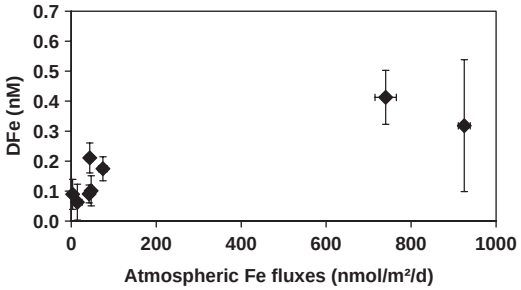

Fig. 7. Mean DFe concentration versus total atmospheric deposition fluxes of soluble Fe (aerosol sample A3 has been excluded from this analysis).

In the parts of our study area where high atmospheric deposition occurred (21.1–16.1°N and 15.9–10.6°N), we estimated residence times for DFe in the surface waters relative to atmospheric inputs. We combined total atmospheric inputs of soluble Fe (dry + wet) and DFe concentrations, assuming a mixed layer depth of 50 ± 20 m along the transect (Robinson et al., 2002). This calculation assumed that the atmospheric deposition we measured was representative in relation to the residence time of water column Fe. These estimations (Table 3) were an upper limit since the other DFe fluxes (advection and diffusion) were not taken into account. Errors were estimated by a Monte Carlo routine (Anderson, 1976). Residence times ranged from 17 ± 8 (21.1–16.1°N) to 28 ± 16 d (15.9–10.6°N). They were equivalent to those estimated in the surface waters of the North and equatorial Atlantic (26–55 d; de Baar and de Jong, 2001), suggesting that atmospheric deposition may be one of the dominant sources of Fe to the surface waters in this area. In any case, these values are very short compared to other trace elements such as aluminium and manganese (4.5–13 and 9.6–11 yr, respectively, Jickells, 1999; Vink and Measures, 2001), and reflect a rapid removal of DFe from the surface waters either via biological uptake, passive adsorption and/or aggregation. Wu et al. (2001) showed that only a small fraction of the sea-surface DFe in regions with high atmospheric Fe supply existed in the size-fraction lower than 0.02 μm. Colloidal aggregation may then be the main factor controlling Fe removal from the surface waters (Wu et al., 2001). If Fe introduced by eolian deposition is mainly colloidal, it may be

Table 3
Estimated residence times of DFe (in days) in the surface waters of the North-Eastern Atlantic Ocean relative to atmospheric inputs

Position	Mean sea-surface DFe	Total atmospheric fluxes	Residence times
21.1–16.1°N	0.32 ± 0.08	925 ± 14	17 ± 8
15.9–10.6°N	0.41 ± 0.13	740 ± 24	28 ± 16

DFe concentrations are in nM and fluxes are in nmol $m^{-2} d^{-1}$.

less bioavailable, provided the Fe uptake is limited by diffusion (Wu et al., 2001). Biological uptake of Fe may also be reduced if other factors are limiting the primary production. In the central Atlantic, it was shown that phosphorus was unlikely to be a limiting resource for planktonic growth (Cañellas et al., 2000), whereas the system was severely nitrogen limited (Augusti et al., 2001).

In the other parts of our transect, particularly in the south Atlantic, atmospheric Fe deposition fluxes were much lower and could not be used alone for estimation of residence times. For a better estimation of residence times, we would need detailed vertical profiles of DFe, T, and S to calculate vertical advective and diffusive fluxes (Vidal et al., 1999; Planas et al., 1999). However, the very low concentrations of DFe in the South Atlantic suggested that Fe might be a (co-)limiting factor of primary production.

Acknowledgements

The authors wish to thank the principal scientist Saad El Dine El Naggar, together with the officers and crew of the R.V. *Polarstern*. We are also thankful to the reviewers for their helpful comments. This work was funded by the EU programme IRONAGES (Iron-Resources and Oceanic Nutrients-Advancement of Global Environment Simulations, EVK2-1999-00031).

References

Anderson, G.M., 1976. Error propagation by the Monte-Carlo method in geochemical calculations. Geochimica et Cosmochimica Acta 40, 1533–1538.

Augusti, A., Duarte, C.M., Vaqué, D., Hein, M., Gasol, J.M., Vidal, M., 2001. Food-web structure and elemental (C, N, and P) fluxes in the eastern tropical North Atlantic. Deep-Sea Research II, 48, 2295–2321.

Baith, K., Lindsay, R., Fu, G., McClain, C.R., 2001. SeaDAS: a data analysis system for ocean colour satellite sensors. EOS, Transactions of American Geophysical Union 82 (18), 102.

Barrie, L.A., 1992. Scavenging ratios: black art or a useful scientific tool? In: Slinn, G., Schwartz, S. (Eds.), Fifth International Conference on Precipitation Scavenging and Atmospheric Exchange Processes. Hemisphere, Washington, DC, pp. 403–419.

Bates, T.S., Quinn, P.K., Coffman, D.J., Johnson, J.E., Miller, T.L., Covert, D.S., Wiedensohler, A., Leinert, S., Nowak, A., Neususs, C., 2001. Regional, physical and chemical properties of the marine boundary layer aerosols across the Atlantic during Aerosols 99: An overview. Journal of Geophysical Research—Atmospheres 106 (D18), 20767–20782.

Bowie, A.R., Whitworth, D.J., Achterberg, E.P., Mantoura, R.F.C., Worsfold, P.J., 2002. Biogeochemistry of Fe and other trace elements (Al, Co, Ni) in the upper Atlantic Ocean. Deep-Sea Research I 49 (4), 605–636.

Bowie, A., Achterberg, E., Blain, S., Boye, M., Croot, P., de Baar, H.J.W., Laan, P., Sarthou, G., Worsfold, P.J. Shipboard intercomparison of dissolved iron in surface waters along a north–south transect of the tropical Atlantic Ocean. Marine Chemistry, in press.

Boyd, P.W., Watson, A.J., Law, C.S., Abraham, E.R., Trull, T., Murdoch, R., Bakker, D.C.E., Bowie, A., Buesseler, K.O., Chang, H., Charette, M., Croot, P., Downing, K., Frew, R., Gall, M., Hadfield, M., Hall, J., Harvey, M., Jameson, G., LaRoche, J., Liddicoat, M., Ling, R., Maldonado, M.T., McKay, R.M., Nodder, S., Pickmere, S., Pridmore, R., Rintoul, S., Safi, K., Sutton, P., Strzepek, R., Tanneberger, K., Turner, S., Waite, A., Zeldis, J., 2000. A mesoscale phytoplankton bloom in the polar Southern Ocean stimulated by iron fertilization. Nature 407, 695–702.

Bruland, K.W., Orians, K.J., Cowen, J.P., 1994. Reactive trace metals in the stratified central North Pacific. Geochimica et Cosmochimica Acta 58, 3171–3182.

Bruland, K.W., Rue, E.L., Smith, G.J., 2001. Iron and macronutrients in California coastal upwelling regimes: Implications for diatom blooms. Limnology and Oceanography 46 (7), 1661–1674.

Bucciarelli, E., Blain, S., Tréguer, P., 2001. Iron and manganese in the wake of the Kerguelen Islands (Southern Ocean). Marine Chemistry 73 (1), 21–36.

Cañellas, M., Agusti, S., Duarte, C.M., 2000. Latitudinal variability in phosphate uptake in the central Atlantic. Marine Ecology Progress Series 194, 283–294.

Carlson, T.N., Prospero, J.M., 1972. The large-scale movement of Saharan air outbreaks over the northern equatorial Atlantic. Journal of Applied Meteorology 11, 283–297.

Coale, K.H., Johnson, K.S., Fitzwater, S.E., Gordon, R.M., Tanner, S., Chavez, F.P., Ferioli, L., Sakamoto, C., Rogers, P., Millero, F., Steinberg, P., Nightingale, P., Cooper, D., Cochlan, W.P., Landry, M.R., Constantinou, J., Rollwagen, G., Trasvina, A., Kudela, R., 1996. A massive phytoplankton bloom induced by an ecosystem scale iron fertilization experiment in the equatorial Pacific Ocean. Nature 383, 495–501.

de Baar, H.J.W., de Jong, J.T.M., 2001. Distributions, sources and sinks of iron in seawater. In: Turner, D.R., Hunter, K.A. (Eds.), Biogeochemistry of Fe in Seawater. SCOR/IUPAC. Chichester, pp. 123–253.

de Jong, J.T.M., den Das, J., Bathman, U., Stoll, M.H.C., Kattner, G., Nolting, R.F., de Baar, H.J.W., 1998. Dissolved iron at subnanomolar levels in the Southern Ocean as determined by ship-board analysis. Analytica Chimica Acta 377, 113–124.

Dierssen, H., Balzer, W., Landing, W.M., 2001. Simplified synthesis of an 8-hydroxyquinoline chelating resin and a study of trace metal profiles from Jellyfish Lake. Palau 73 (3–4), 173–192.

Duce, R.A., Tindale, N.W., 1991. Atmospheric transport of iron and its deposition in the ocean. Limnology and Oceanography 36, 1715–1726.

Duce, R.A., Liss, P.S., Merrill, J.T., Atlas, E.L., Buat-Ménard, P., Hicks, B.B., Miller, J.M., Prospero, J.M., Arimoto, R., Church, T.M., Ellis, W., Galloway, J.N., Hansen, L., Jickels, T.D., Knap, A.H., Reinhardt, K.H., Schneider, B., Soudine, A., Tokos, J.J., Tsunogai, S., Wollast, R., Zhou, M., 1991. The atmospheric input of trace species to the world ocean. Global and Biogeochemical Cycles 5 (3), 193–259.

Elliott, W.P., Reed, R.K., 1984. A climatological estimate of precipitation for the world ocean. Journal of Climate and Applied Meteorology 23 (3), 434–439.

Gledhill, M., Van den Berg, C.M.G., 1994. Determination of complexation of iron(III) with natural organic complexing ligands in seawater using cathodic stripping voltammetry. Marine Chemistry 47, 41–54.

Grgic, I., Dovzan, A., Bercic, G., Hudnik, V., 1998. The effect of atmospheric organic compounds on the Fe-catalysed S(IV) autoxidation in aqueous solution. Journal of Atmospheric Chemistry 29, 315–337.

Guieu, C., Bozec, Y., Blain, S., Ridame, C., Sarthou, G., Leblond, N., 2002. Impact of high Saharan dust inputs on dissolved iron distributions in the Mediterranean Sea. Geophysical Research Letters 29 (19), 1911.

Helmers, E., Schrems, O., 1995. Wet deposition of metals to the tropical North and the South Atlantic Ocean. Atmospheric Environment 29 (18), 2475–2484.

Hoffmann, P., Dedik, A.N., Esling, J., Weinbruch, S., Weber, S., Sinner, T., Gutlich, P., Ortner, H.M., 1996. Speciation of iron in atmospheric aerosol samples. Journal of Aerosol Science 27 (2), 325–337.

Jickells, T., 1999. The inputs of dust derived elements to the Sargasso Sea: a synthesis. Marine Chemistry 68, 5–14.

Jickells, T., Spokes, L.J., 2001. Atmospheric iron inputs to the oceans. In: Turner, D.R., Hunter, K.A. (Eds.), The biogeochemistry of Iron in seawater. SCOR-IUPAC, Baltimore, pp. 85–121.

Johansen, A.M., Siefert, R.L., Hoffmann, M.R., 2000. Chemical composition of aerosols collected over the tropical North Atlantic Ocean. Journal of Geophysical Research 105 (D12), 15277–15312.

Johnson, K.S., Gordon, R.M., Coale, K.H., 1997. What controls dissolved iron concentrations in the world ocean? Marine Chemistry 57, 137–161.

Kim, G., Church T, M., 2002. Wet deposition of trace elements and radon daughter systematics in the South and equatorial Atlantic atmosphere. Global Biogeochemical Cycles 16 (3), 1046.

Lim, B., Jickells, T.D., 1990. Dissolved, particulate and acid-leachable trace metal concentrations in North Atlantic precipitation collected on the global change expedition. Global Biochemical Cycles 4 (4), 445–458.

Liu, X., Millero, F.J., 2002. The solubility of iron in seawater. Marine Chemistry 77 (1), 43–54.

Losno, R., Bergametti, G., Carlier, P., 1992. Origin of atmospheric particulate matter over the North Sea and the Atlantic Ocean. Journal of Atmospheric Chemistry 15 (3/4), 333–352.

Loÿe-Pilot, M.D., Martin, J.-M., 1996. Saharan dust input to the Western Mediterranean: an eleven years record in Corsica. In: Guerzoni, S., Chester, R. (Eds.), The Impact of Desert Dust Across the Mediterranean. Kluwer Academic Publishers, Dordrecht, pp. 191–199.

Maenhaut, W., Salma, I., Cafmeyer, J., Annegarn, H.J., Andrea, M.O., 1996. Regional atmospheric aerosol composition and sources in the eastern Transvaal, South Africa, and impact of biomass burning. Journal of Geophysical Research—Atmosphere 101 (D19), 23631–23650.

Maring, H., Savoie, D.L., Izaguirre, M.A., McCormick, C., Arimoto, R., Prospero, J.M., Pilinis, C., 2000. Aeorosol physical and optical properties and their relationship to aerosol composition in the free troposphere at Izana, Tenerife, Canary Island, during July 1995. Journal of Geophysical Research—Atmosphere 105 (D11), 14677–14700.

Martin, J.H., Coale, K.H., Johnson, K.S., Fitzwater, S.E., Gordon, R.M., Tanner, S.J., Hunter, C.N., Elrod, V.A., Nowiicki, J.L., Coley, T.L., Barber, R.T., Lindley, S., Watson, A.J., Van Scoy, K., Law, C.S., Liddicoat, M.I., Ling, R., Stanton, T., Stockel, J., Collins, C., Anderson, A., Bidigare, R., Ondrusek, M., Latasa, M., Millero, F.J., Lee, K., Yao, W., Zhang, J.Z., Friederich, G., Sakamoto, C., Chavez, F., Buck, K., Kolber, Z., Greene, R., Falkowski, P., Chisholm, S.W., Hoge, F., Swift, R., Yungel, J., Turner, S., et al., 1994. Testing the iron hypothesis in ecosystems of the equatorial Pacific Ocean. Nature 371, 123–129.

Martin, J.H., Fitzwater, S.E., 1988. Iron deficiency limits phytoplankton growth in the north-east Pacific subarctic. Nature 331, 341–343.

Martin, J.H., Fitzwater, S.E., Gordon, R.M., 1990. Iron deficiencies limits plankton growth in Antarctic waters. Global Biogeochemical Cycles 4, 5–12.

Moulin, C., Lambert, C.E., Dulac, F., Dayan, U., 1997. Control of atmospheric export of dust from North Africa by the North Atlantic oscillation. Nature 387, 691–694.

Obata, H., Karatani, H., Nakayama, E., 1993. Automated determination of iron in seawater by chelating resin concentration and chemiluminescence. Analytical Chemistry 65, 1524–1528.

Piketh, S.J., Annegarn, H.J., Tyson, P.D., 1999. Lower-tropospheric loadings over South Africa: the relative contributions of aeolian dust, industrial emissions and

biomass burning. Journal of Geophysical Research—Atmosphere 104 (D1), 1597–1607.

Planas, D., Agusti, S., Duarte, C.M., Granata, T.C., 1999. Nitrate uptake and diffusive nitrate supply in the Central Atlantic. Limnology and Oceanography 44 (1), 116–126.

Powell, R.T., Donat, J.R., 2001. Organic complexation and speciation of iron in the South and Equatorial Atlantic. Deep-Sea Research II 48 (13), 2877–2893.

Powell, R.T., King, D.W., Landing, W.M., 1995. Iron distributions in the surface waters of the south Atlantic. Marine Chemistry 50 (1–4), 13–20.

Prospero, J.M., Uematsu, M., Savoie, D.L., 1989. Mineral aerosol transport to the Pacific Ocean. In: Riley, J.P., Chester, R., Duce, R.A. (Eds.), Chemical Oceanography, Academic Press, London, pp. 187-218.

Rädlein, N., Heumann, K.G., 1992. Trace analysis of heavy metals in aerosols over the Atlantic Ocean from Antarctica to Europe. International Journal of Environmental Analytical Chemistry 48 (2), 127–150.

Rädlein, N., Heumann, K.G., 1995. Size-fractionated impactor sampling of aerosols particles over the Atlantic Ocean from Antarctica to Europe as a methodology for source identification of Cd, Pb, Ti, Ni, Cr, and Fe. Fresenius Journal of Analytical Chemistry 352 (7–8), 748–755.

Raes, F., van Dingenen, R., Vignati, E., Wilson, J., Putaud, J.P., Seinfeld, J.H., Adams, P., 2000. Formation and cycling of aerosols in the global troposphere. Atmospheric Environment 35 (25), 4215–4240.

Robinson, C., Serrert, P., Tilstone, G., Teira, E., Zubkov, M.V., Rees, A.P., Woodward, E.M.S., 2002. Plankton respiration in the Eastern Atlantic Ocean. Deep-Sea Research II 49, 787–813.

Rue, E.L., Bruland, K.W., 1995. Complexation of iron(III) by natural organic ligands in the Central North Pacific as determined by a new competitive ligand equilibration/adsorptive cathodic stripping voltammetric method. Marine Chemistry 50 (1–4), 117–138.

Rue, E.L., Bruland, K.W., 1997. The role of organic complexation on ambient iron chemistry in the equatorial Pacific Ocean and the response of a mesoscale iron addition experiment. Limnology and Oceanography 42 (5), 901–910.

Sarthou, G., Jeandel, C., 2001. Seasonal variations of iron concentrations in the Ligurian Sea and iron budget in the Western Mediterranean Sea. Marine Chemistry 74 (2–3), 115–129.

Siedler, G., Zangenberg, N., Onkeb, R., Morlière, A., 1992. Seasonal changes in the tropical Atlantic circulation: observation and simulation of the guinea dome. Journal of Geophysical Research 97, 703–715.

Smetacek, V., 2001. EisenEx: international team conducts iron experiment in Southern Ocean. USJGOFS Newsletter January, 11–14.

Spokes, L.J., Jickells, T.D., 1996. Factors controlling the solubility of aerosol trace metals in the atmosphere and on mixing into seawater. Aquatic Geochemistry 1, 355–374.

Spokes, L.J., Jickells, T.D., Lim, B., 1994. Solubilisation of aerosol trace metals by cloud processing; a laboratory study. Geochimica et Cosmochimica Acta 58 (15), 3281–3287.

Sultan, B., Janicot, S., 2000. Abrupt shift of the ITCZ over West Africa and intra-seasonal variability. Geophysical Research Letters 27 (20), 3353–3356.

Swap, R., Ulanski, S., Cobbett, M., Garstand, M., 1996. Temporal and spatial characteristtics of Saharan dust outbreaks. Journal of Geophysical Research 101, 4205–4220.

Tchernia, P., 1980. Descriptive Regional Oceanography. Pergamon Press, New York, pp. 1–253.

Torres-Padron, M.E., Gelado-Caballero, M.D., Collado-Sanchez, C., Siruela-Matos, V.F., Cardona-Castellano, P.J., Hernandez-Brito, J.J., 2002. Variability of dust inputs to the CANIGO zone. Deep-Sea Research II 49 (17), 3455–3464.

Tyson, P.D., Garstang, M., Swap, R., Kallberg, P., Edwards, M., 1996. An air transport climatology for subtropical southern Africa. International Journal of Climatology 16, 265–291.

Vidal, M., Duarte, C.M., Agusti, S., 1999. Dissolved organic nitrogen and phosphorus pools and fluxes in the central Atlantic Ocean. Limnology and Oceanography 44 (1), 106–115.

Vink, S., Measures, C.I., 2001. The role of dust deposition in determining surface water distributions of Al and Fe in the South West Atlantic. Deep-Sea Research II 48 (13), 2787–2809.

Völkening, J., Heumann, K.G., 1990. Heavy metals in the near surface aerosols over the Atlantic from 60-degrees South to 54-degrees North. Journal of Geophysical Research—Atmosphere 95 (D12), 20623–20632.

Wedepohl, K.H., 1995. The composition of the continental crust. Geochimica et Cosmochimica Acta 59, 1217–1232.

Wu, J., Luther III, G.W., 1994. Size-fractionated iron concentrations in the water column of the Northwest Atlantic Ocean. Limnology and Oceanography 39, 1119–1129.

Wu, J., Boyle, E., Sunda, W., Wen, L.-S., 2001. Soluble and colloidal iron in the oligotrophic North Atlantic and North Pacific. Science 293, 847–849.

Zhu, X., Prospero, J.M., Savoie, D.L., Millero, F.J., Zika, R.G., Saltzman, E.S., 1993. Photoreduction of iron(III) in marine mineral aerosol solutions. Journal of Geophysical Research 98, 9039–9046.